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THE NATURE OF INDIVIDUAL RADIOACTIVE PARTICLES
VI. FALLOUT PARTICLES FROM A
TOWER SHOT, OPERATION REDWING

Research and Development Technical Report USNRDL-TR-208
NS 081-001

2 December 1957

by

RICHARD COLE

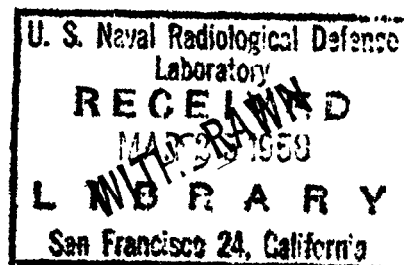


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U.S. NAVAL RADIOLOGICAL DEFENSE LABORATORY

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THE NATURE OF INDIVIDUAL RADIOACTIVE PARTICLES
VI. FALLOUT PARTICLES FROM A
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Chemistry—
Radiation and Radiochemistry

Technical Objective
AW-7

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Encl: (1) U.S. Naval Radiological Defense Laboratory Report USNRDL-TR-208
entitled "The Nature of Individual Radioactive Particles.
VI. Fallout Particles from a Tower Shot, Operation REDWING"

1. Enclosure (1) is the sixth in a series of reports describing the properties of radioactive fallout particles originating from the detonation of nuclear weapons in various types of physical environment. Studies were made of the structure and composition of and the distribution of radioactivity within the fallout particles collected following a tower shot during Operation REDWING at the Eniwetok Proving Grounds. Techniques utilizing the petrographic microscope and X-ray diffraction analysis were employed.

2. The most common type of fallout particle found was formed by the interaction of the vaporized iron and radioactive elements with molten calcium oxide derived from the coral sand carried up into the fireball by atmospheric turbulence. A second type consisted almost wholly of radioactive iron oxide apparently formed by the melting and oxidation of part of the steel tower. A third type consisted of unmelted coral sand grains which had small, black, radioactive spheres adhering to their surfaces.


for PAUL C. TOMPKINS
By direction

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ABSTRACT

Studies were made of the structure and composition of, and the distribution of radioactivity within, the fallout particles resulting from a tower shot in Operation REDWING. Techniques utilizing the petrographic microscope and X-ray diffraction analysis were employed. Three types of fallout particle were found. The most common type was formed by the interaction of the vaporized iron and radioactive elements with molten calcium oxide derived from the coral sand which was carried up into the fireball by atmospheric turbulence. These particles consisted of an outer zone of radioactive dicalcium ferrite surrounding an inactive residual core of altered calcium oxide. The second type consisted almost wholly of radioactive iron oxide and apparently was formed by the melting and oxidation of part of the steel tower. The third type consisted of unmelted coral sand grains which had small, black, radioactive spheres adhering to their surfaces.

This is the sixth in a series of reports on the nature of radioactive fallout particles: Part I, USNRDL-374, 28 November 1952; II, USNRDL-408, 1 July 1953; III, USNRDL-440, 24 February 1954; IV, USNRDL-TR-26, 17 January 1955; V. USNRDL-TR-133, 1 February 1957; all by C. E. Adams.

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SUMMARY

The Problem

This is the sixth in a series of reports devoted to the study of radioactive fallout particles originating from the detonations of nuclear weapons in various types of physical environment. The fallout particles described here were collected following a tower shot in Operation REDWING, at the Eniwetok Proving Grounds. As in previous fallout particle studies, the chemical composition and structure of the particles and the distribution of radioactivity within them was desired.

Findings

The chemical compositions and structures of the particles were determined by studying thin sections of the particles under the petrographic microscope. The distribution of radioactivity within the particles was found by making radioautographs of the thin sections. Determinations of the chemical compositions were also made by X-ray diffraction analysis.

Three types of radioactive fallout particles were found. The most common type was composed of radioactive dicalcium ferrite surrounding an inner core of altered calcium oxide. These particles were formed by the interaction of the iron and radioactive elements condensing in the cooling fireball with drops of melted calcium oxide derived from the coral sand which was carried up into the fireball by atmospheric turbulence. A second type consisted wholly of radioactive iron oxide and was formed by the melting but incomplete vaporization of the steel tower with subsequent oxidation of the liquid iron drops. The third type consisted of unmelted coral sand grains with small, black, radioactive spheres adhering to their surfaces. The small, black spheres were presumably formed by the direct condensation of vaporized material in the fireball.

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ADMINISTRATIVE INFORMATION

The investigation reported was done under Bureau of Ships Project No. NS 081-001, Technical Objective AW-7. The work is part of the technical program for the Department of the Army established between Office, Chief of Research and Development, Department of the Army, and the Bureau of Ships (joint agreement, 23 November 1955).

The work is described, as Program 1, Problem 1, in this laboratory's Preliminary Presentation of USNRDL Technical Program for FY 1957, of February 1956.

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INTRODUCTION

A study has been made of the chemical composition, structure and distribution of radioactivity in the radioactive fallout particles resulting from a moderate-yield tower shot in Operation REDWING at the Eniwetok Proving Grounds.

To collect samples of the radioactive fallout, large, open trays were placed on several of the islands which extended along the atoll for a few miles on each side of the shot point. On only one of these islands, approximately ten miles from the shot point, was fallout sufficiently heavy to provide abundant samples of the fallout particles. The trays from this island were picked up, secured for transportation and shipped to this laboratory.

In order to study the radioactive fallout particles it was necessary first to separate them from the inactive sand grains which had been swept into the open trays by surface winds. This was done by removing all the particles from the trays and sprinkling them onto the backs of pieces of single-coated X-ray sheet film. Just before dispersing the particles onto the film, the film had been sprayed with a thin layer of liquid plastic which, after hardening, held the scattered particles onto the film. After an exposure of a few hours the films were developed and fixed by brushing the appropriate solutions onto the emulsion side of the film, with care not to disturb the particles on the other side. The radioactive particles were easily identified by the halo of darkened film surrounding them.

Many of the radioactive particles were then removed from the films and mounted in small plastic pellets which were then ground into thin sections with standard petrological techniques. These sections, which were cut through the center of each particle, were about 30 μ thick. By studying these thin sections under the petrographic microscope it was possible to identify the transparent compounds and to observe the structure of the fallout particles. The opaque compounds in the particles were identified by X-ray diffraction analysis. The distribution of radioactivity within the particles was found by radioautographing the thin sections with Eastman NTB stripping film.

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DESCRIPTION OF THE FALLOUT PARTICLES

Three types of radioactive fallout particles were observed. The most abundant particles were spheroidal, dull black, cracked and veined with white crystalline material and varied in size from about 1/2 to 1 mm in diameter. Examination of the thin sections of these particles revealed that they had a diffuse, central core of what had originally been calcium oxide but which had been subsequently converted to calcium carbonate and calcium hydroxide. Surrounding this central core was a thick zone of black, opaque material which was identified as dicalcium ferrite ($2 \text{CaO} \cdot \text{Fe}_2\text{O}_3$). The white material filling the veins in the particles, and sometimes occurring as surface coatings, was found to be mixtures of calcite and vaterite. Calcite and vaterite are different crystal modifications of calcium carbonate and differ from the crystal form of the calcium carbonate in the original coral which is largely aragonite. The radioactivity in this type of particle was always associated with the areas of dicalcium ferrite; the calcium compounds in the core and veins were inactive (see Fig. 1).

The second most abundant type of particle was spherical, black with a glossy luster, magnetic, and about 1/4 mm in diameter. These particles were found to consist of magnetite (Fe_3O_4) and some hematite (Fe_2O_3). The radioactivity was distributed more or less homogeneously throughout the volumes of the particles (see Fig. 2).

The third type of particle was found comparatively rarely. These particles were white and irregular in shape, and had the appearance of coral sand grains. Examination of the thin sections showed that these particles were composed either of unaltered coral or of calcium hydroxide with a thin outer coating of calcium carbonate. Adhering to the surfaces of these particles were many small, black spheres ranging in size from about 10 μ in diameter to submicroscopic. Radioautographs revealed that these small black spheres were the carriers of the radioactivity and that the large calcium carbonate or hydroxide particles were inactive (see Fig. 3).

ORIGIN OF THE FALLOUT PARTICLES

The fallout particles of the first type described above were formed by the interaction of the condensing material, which had been vaporized in the fireball, with the surface material swept up into the cooling fireball by atmospheric turbulence. The vaporized material consisted predominantly of iron derived from the steel tower and some calcium oxide derived from the coral sand.

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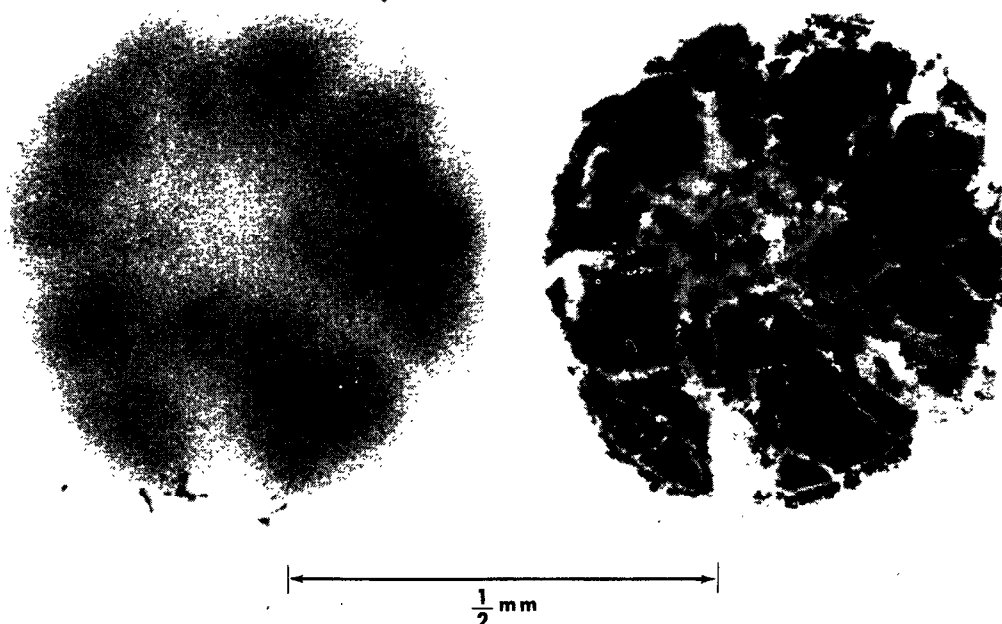


Fig. 1 Thin Section and Radioautograph of a Fallout Particle. It consists of an outer zone of dicalcium ferrite (black areas) and an inner core and veins of calcium carbonate and calcium hydroxide (gray areas). The radioactivity is associated with the iron-rich part of the particles; the calcium compounds in the core and veins are inactive.

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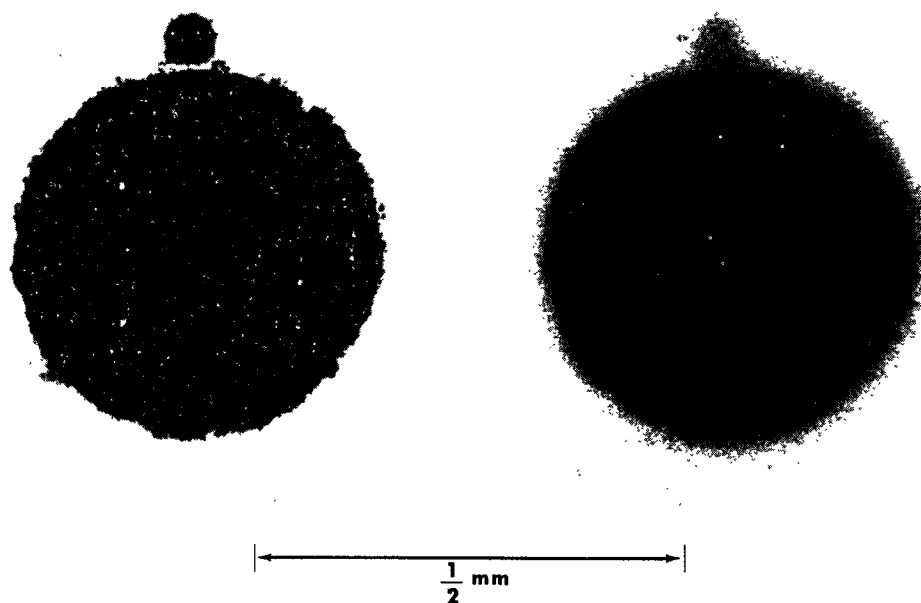


Fig. 2 Thin Section and Radioautograph of a Fallout Particle. It consists of iron oxide (magnetite). The radioactivity is distributed homogeneously throughout the particle.

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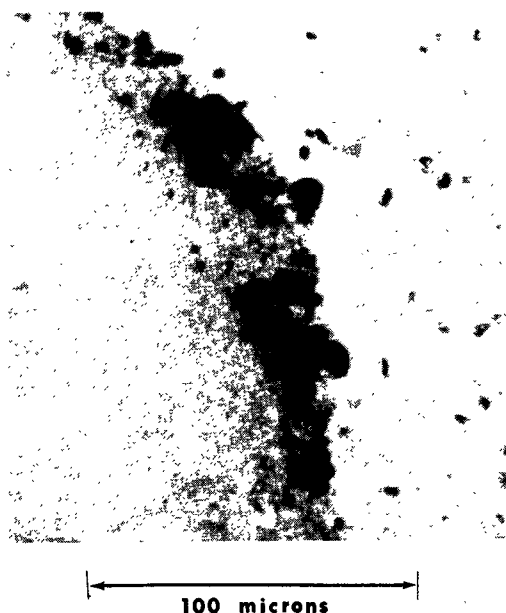


Fig. 3 Part of a Thin Section of a Fallout Particle. It consists of calcium hydroxide with a thin surface coating of calcium carbonate. Adhering to the surface of the particle are many, small, black, radioactive spheres. The calcium compounds are inactive.

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The surface material swept up into the fireball was converted to liquid calcium oxide drops by decarbonation and melting of the coral sand grains. The iron in the fireball collected on the surfaces of these calcium oxide drops either by direct condensation from the vapor state or by the impaction of small iron oxide drops. These small iron oxide drops were formed either by direct condensation from the vapor state or by the melting and oxidation of the steel tower.

The iron reacted in the oxidizing atmosphere with the calcium oxide to form the dicalcium ferrite. In most cases a residual core of unaltered calcium oxide was left. The fission products condensed with the iron, thereby making the iron-rich areas of the particles radioactive. After cooling, the calcium oxide in the central area was carbonated and hydrated by the carbon dioxide and water vapor in the air. During this process there was a considerable expansion which cracked the particles and formed fissures and veins which filled with the calcium compounds diffusing from the interior.

Particles of the second type, consisting almost wholly of iron oxide, were probably formed by the oxidation of small liquid iron drops formed by the melting but incomplete vaporization of the steel tower. The iron drops would be readily oxidized in the heated air, and the condensing materials, iron as well as the radioactive elements, would be dissolved and distributed throughout the liquid drop.

There also is the possibility that the tower was completely vaporized and that the iron oxide particles were formed by direct condensation of the iron from the vapor state. However, Stewart¹ has made an analysis of the growth of particles by a process of condensation and coagulation from the fireball of a tower shot in which his assumed conditions were much the same as those occurring at the shot studied here. He concluded that the modal diameter of the iron oxide particles would be only 0.1 μ . From his expression for the particle size distribution resulting from the condensation-coagulation process it can be inferred that it would be highly improbable that the particles could attain a diameter of a few hundred microns in the few seconds available for growth.

The third type of fallout particle described above was formed by coral sand grains entering the fireball at a comparatively late time and collecting on their surfaces the small, black, radioactive spheres. These spheres were too small to analyze but they were presumably formed by direct condensation of the vaporized material in the fireball. The coral sand grains were usually heated sufficiently to decarbonate them (800 to 900° C) but not sufficiently to melt the resulting oxide (2570° C). They were subsequently hydrated to calcium hydroxide by water vapor and a thin outer layer of calcium carbonate was formed by the carbon dioxide in the atmosphere. In some cases the sand grains were not heated sufficiently to decarbonate them and they still retained the structure and composition of the original coral.

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DISCUSSION

In the description of the formation of the fallout particles given above little attention has been paid to the behavior of the radioactive elements themselves. These radioactive elements are present in such small quantities that they may be considered as trace constituents and do not affect the main course of fallout particle formation. However, as an initial step in a more detailed study of the condensation of the vaporized radioactive elements, it was desired to know something about the temperature range in the fireball at which they condense. The general process of fallout particle formation described in this report, in which melted soil particles act as collecting surfaces for the radioactive elements in the fireball, had been previously observed at a tower shot at the Nevada Test Site.² The soil at the Nevada Test Site consists mostly of silicate minerals which melt at comparatively low temperatures (approximately 1200 to 1700° C). The fallout particles in this case consisted of transparent glass drops with an outer zone of intensely colored, radioactive glass formed by solution of the iron from the tower and the radioactive elements in the fused soil drops.

With this process in mind, an experiment was devised in which a series of high-melting tracer minerals were to be introduced into the natural soil surrounding the tower for a shot of appropriate size. It was anticipated that these tracer minerals would be drawn up into the cooling fireball and act as collecting surfaces for the condensing radioactive elements. By using tracer minerals which had considerably higher melting points than the Nevada soil minerals it was hoped that these higher-melting species would enter the cooling fireball and collect some of the condensing radioactive elements but would not be heated sufficiently to melt. By collecting fallout particles from this shot and noting which of the tracer minerals had collected radioactivity but had not melted, it would be possible to tell approximately at what temperature condensation was taking place.

Accordingly, 14 tons of tracer minerals were distributed around the tower of the shot at Eniwetok whose fallout is described in this report. The pertinent data concerning these minerals are given in Table 1.

These minerals had been ground and sieved and only material whose particle size was about 0.8 mm in diameter or less was used. The minerals were scattered lightly over the top of the ground in areas extending from the base of the tower out to about 150 yards. The natural coral soil at Eniwetok furnished calcium oxide, which melts at 2570° C, as the highest-melting member of the series.

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TABLE 1
Tracer Mineral Data

Tracer Mineral	Melting or Disintegration Point (°C)	Amount Used (tons)
Microcline (KAlSi_3O_8)	1170-1450	3
Barite (BaSO_4)	1580	3
Quartz (SiO_2)	1710	3
Rutile (TiO_2)	1840	2
Zircon (ZrSiO_4)	2550	3

At shot time the winds were unfavorable so that usable amounts of fallout were collected at only one island station. As has been described above, the temperature at which the fallout particles received their radioactive contamination exceeded the melting point of calcium oxide so that all the lower melting minerals were either completely melted or vaporized. None of the tracer minerals were recovered in an unaltered form. However, spectrographic analysis revealed the presence of certain characteristic elements from the tracer minerals in the fallout particles. In order to determine the fate of the tracer minerals, a 125-mg sample of fallout particles was accumulated and a chemical analysis was made for their iron and calcium content, as well as for certain of the elements found in the tracer minerals but not found in appreciable quantities in the coral sand or in the tower or associated structures. The weight of the iron in the tower is known and, by assuming that the composition of the analyzed fallout particles is representative of the composition of the fireball material, it is possible to compute the weight of the coral sand and of the tracer minerals which entered the fireball and were involved in fallout particle formation. The results of these computations are given in Table 2.

While these figures can be considered as only approximations due to the inaccuracies in analyzing for such small amounts of material and due to the error inherent in the assumption concerning the representativeness of the sample, nevertheless it can be seen that a substantial fraction of the tracer minerals and about 264 tons of coral sand entered into fallout particle formation.

TABLE 2

Fallout Particle Composition

Element	Weight in Fallout Sample (mg)	Weight in Fireball (tons)	Computed Weight of Original Minerals in Fireball (tons)
Fe	53.7	165	-
Ca	34.3	106	264 (CaCO ₃)
Ba	2.21	6.8	11.6 (BaSO ₄)
Ti	0.45	1.4	2.3 (TiO ₂)
Zr	0.25	0.77	1.5 (ZrSiO ₄)
Al	0.16	0.50	3.0 ^(a) (KAlSi ₃ O ₈)

(a) Corrected for weight of Al in tower.

The question arises as to how much of this material, especially of the large quantity of coral sand, was vaporized and how much only melted. It is known from an examination of the thin sections that part of the calcium oxide in the fallout particles occurs as an inactive, residual core and was originally melted but not vaporized. However, most of the calcium oxide in the particles occurs in combination with the iron oxide and it is not possible to tell whether this part was originally vaporized and condensed with the iron or whether it was part of the original liquid drop and merely dissolved and reacted with the condensing iron. The existence of radioactive fallout particles consisting essentially of iron oxide (magnetite) indicates that relatively little of the calcium oxide was vaporized. If there had been appreciable amounts of the calcium oxide vapor it would presumably have condensed onto the iron oxide drops as well as onto the calcium oxide drops. The fact that X-ray diffraction analysis detected no calcium compounds in the iron oxide particles shows that the amount of vaporized calcium oxide was small.

A qualitative spectrographic analysis was made of a sample of the iron oxide fallout particles and the characteristic elements from the tracer minerals were found in them. As none of the tracer minerals were ever found embedded in any of the particles, their occurrence suggests they were probably vaporized to some extent and condensed onto the particles. It is also possible that some of the small tracer mineral particles were drawn into the fireball and impacted on and dissolved in the liquid oxide drops.

U N C L A S S I F I E D

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2. Adams, C.E., and Wittman, J.P. The Nature of Individual Radioactive Particles, Part III (short title). U. S. Naval Radiological Defense Laboratory Research and Development Report USNRDL-440, 24 February 1954, CLASSIFIED.
3. The Effects of Nuclear Weapons. U. S. Atomic Energy Commission, available through U. S. Government Printing Office, Washington 25, D. C., June 1957, p. 285.

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